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Cyclotron Resonance of the Polaron.

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CYCLOTRON RESONANCE OF THE POLARON

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ABSTRACT

Frohlich has developed a Hamiltonian to describe the motion of a single Bloch conduction electron in an ionic solid, including the interaction of the electron with the polarization field which the electron induces in the solid. The low-lying energy states of Frohlich's Hamiltonian are descriptive of a single particle, called the polaron, characterized by a self-energy and an effective mass. The polaron may be regarded as a composite entity consisting of the Bloch electron together with its accompanying polarization field. In its dimensionless form, the Hamiltonian and, therefore, also the self-energy and effective mass, are functions of a single parameter, called the polaron coupling constant, which measures the strength with which the electron interacts with the polarization field of the solid. The polaron problem is of interest not only because of its intrinsic value to solid state physics, but also because it provides a relatively simple but realistic example problem of the quantum mechanical description of a particle interacting with its self-produced, quantized field. Therefore, the coupling constant is regarded as an arbitrary parameter.

Frohlich's Hamiltonian is derived and discussed and research literature on the polaron problem is selectively reviewed. The

motion of a polaron in a magnetic field is then considered. It is shown that the self-energy and effective mass as defined by Frohlich for a free polaron also characterize the low-lying energy states of a polaron in a weak magnetic field exactly, regardless of the strength of the polaron coupling, and that (to first order in the magnetic field) a cyclotron resonance experiment would measure the Frohlich polaron mass. In the case of weak coupling, the cyclotron motion of a polaron may be simply described by the use of perturbation theory.

Trial state functions are proposed for the low-energy states of a strongly coupled polaron in a weak magnetic field. These trial state functions are constructed on the basis of a refinement of the intuitive picture of a composite particle with fixed structure whose center behaves as a point charge in the magnetic field. The resultant trial state functions display many of the features which must be possessed by the unknown exact low-lying energy states which they purport to approximate. The polaron self-energy calculated from these trial states functions agrees with well established results. The resultant effective mass agrees with the consensus of results of previous work, but the latter is not established as necessarily correct either on experimental or theoretical grounds. This agreement provides significant independent support to the bulk of previous polaron effective mass calculations.

CHAPTER 1

INTRODUCTION

1. The Definition of the Polaron

If a charged particle of sufficiently low kinetic energy is introduced into a polarizable medium, the charged particle will induce a polarization field which will be concentrated in a region which moves through the medium with the particle. Conceptually, the particle and its accompanying polarization field may be regarded as a composite particle, called a polaron, and described in terms of effective parameters.

In most historical applications, the charged particle has been an electron in an otherwise vacant conduction band of an ionic lattice. The polarization field arises from displacement of the ions from their respective lattice sites and from distortion of the ions resulting from the presence of the electron. In the absence of such effects, F. Bloch[1] has shown that it is frequently possible to regard the conduction electron as behaving much as it does in a vacuum, but with an effective mass (called the Bloch mass) in general different from that of the free electron. With its mass so adjusted, the electron is called a Bloch conduction electron. If conditions are selected so that such an approximation is possible,

then the polaron may be regarded as a Bloch conduction electron together with its accompanying polarization field which the electron induces and with which the electron interacts.

2. The Formulation of the Polaron Problem

The polaron problem was first developed by L. D. Landau[2] in 1933 but was put into its present form by H. Frohlich[3] in 1954. Frohlich formulated the problem in terms of a Hamiltonian written as the sum of three parts:

$$H = H_{BE} + H_p + \alpha^{\frac{1}{2}} H_I . \qquad (1)$$

This Hamiltonian is derived in Chapter 2. The first term, H_{BE}, describes the Bloch conduction electron as a free particle with a scalar mass appropriate for an electron with energy near the bottom of the conduction band of the ionic solid. The second term, H_p, represents the energy of the longitudinal optical vibrational modes of the lattice, all regarded as having the same frequency. All other modes of the ionic solid are neglected, and the solid is treated as a homogeneous, isotropic, continuous medium. The last term, $\alpha^{\frac{1}{2}} H_I$, is the Hamiltonian of interaction of the Bloch conduction electron with the polarization field resulting from the longitudinal optical modes of the ionic solid. The coefficient α is a dimensionless constant, called the polaron coupling constant. As a typical example, α is about 2 for AgBr.

In the study of the polaron problem, some of the quantities of principal interest are the polaron self-energy (the groundstate eigenvalue of H) and the effective mass of the polaron, along with

the results of mobility, cyclotron resonance, and magnetic susceptibility experiments. It is primarily a theoretical description of the cyclotron resonance of the polaron which will be considered in this dissertation.

3. The Significance of the Polaron Problem

The properties of a Bloch conduction electron in an ionic solid have an intrinsic interest, and certain experimental work has been published (see Chapter 5). To a limited extent, the polaron model has been applied to the study of metal-ammonia solutions as well. Aside from these applications, there is considerable interest in the polaron problem as a prototype and testing ground for field-theoretic methods. The Frohlich Hamiltonian represents a single bare particle interacting with a quantized scalar field, where both the particle and the field are treated in a non-relativistic approximation. The problem displays none of the troublesome infinities and divergences which plague other, particularly relativistic, field theories. It has already provided instructive applications of many field-theoretic methods, especially variational approximation methods. Among these, Feynman's variational path integral approximation method[4,5] is particularly noteworthy.

In this dissertation the development of the polaron problem will be in terms of the historical context of a conduction electron in an ionic solid, but because of the extended interest in the polaron problem, the polaron coupling constant, α, will not be

restricted to its range of values appropriate for ionic solids (roughly $.5 < \alpha < 6$).

4. Cyclotron Resonance

A cyclotron resonance experiment is essentially an experiment to determine the effective mass of a charge carrier in a medium. A constant uniform magnetic field is imposed to cause the charge carrier to move (in a classical sense) in circular "Landau orbits," differing in energy by a quantity which depends, among other things, on the mass of the carrier. If an essentially monochromatic electromagnetic field is applied to the medium, resonant power absorption will be observed when the circular frequency of the applied field (times $\hbar$) equals the energy difference between Landau orbits. From the magnetic field strength and the resonant frequency, the mass can be found.

For a magnetic field of arbitrary strength, it would not be justified to presume that the cyclotron motion is associated with the polaron as a whole with the same internal structure as a free polaron. In the appendix it is proved, however, that for any fixed value of the polaron coupling constant, in the weak magnetic field limit, a cyclotron resonance experiment does measure the effective mass of a free polaron at rest. For this reason, this dissertation will be concerned with a description of a polaron only in a limitingly weak magnetic field.

5. Some Conventions

To distinguish various masses, the following conventions will be used. The Bloch mass of the electron will be denoted by m.

The free electron mass will be written as m_e. The effective mass of a free polaron at rest will be called the polaron mass and denoted by m^*. The ratio m^*/m will be written as μ:

$$\mu = m^*/m \ . \tag{2}$$

In the literature, unfortunately, both m^* and μ are referred to as the polaron mass. To avoid misunderstanding, μ will be called the polaron mass ratio. It is convenient to introduce μ since it is a universal function of the polaron coupling constant.

The magnitude of the electron's charge will be denoted e, so that the actual charge on the electron is -e. Rectangular cartesian vectors will be denoted in the usual manner by placing an arrow over the symbol for the vector. In terms of its rectangular cartesian components A_1, A_2, A_3, a vector will be written: $\vec{A} = (A_1, A_2, A_3)$. The letter H will be reserved for Hamiltonian operators, and the symbol ψ_t for trial state functions. The notation $O(a^n)$ denotes a term which is of order n in the parameter a.

The symbol U, subscripted as required, will be reserved for unitary transformations. Where appropriate, the result of applying the transformation U to an operator O will be expressed in the form

$$O \Longrightarrow U^+ O U, \tag{3}$$

and the result of applying U to a state function ψ will be expressed in the form

$$\psi \Longrightarrow U^+ \psi. \tag{4}$$

When this notation is used, the transformation under consideration will be presumed to be understood, and no explicit notation will be used to distinguish original quantities from transformed ones.

CHAPTER 2

DEVELOPMENT OF THE HAMILTONIAN[3,6]

1. The Bloch Approximation[7,8]

Consider the motion of a single electron in an otherwise vacant conduction band of an ionic solid. In the Bloch approximation, the electron is supposed to move in a periodic electrostatic potential produced by the ions of a perfect ionic lattice. The ions are presumed to be static and rigidly attached to their locations in the lattice; they do not react in any way to the presence of the electron.

In the polaron problem, one wishes to treat the band structure of the solid in as simple a manner as possible, concentrating rather on the electron-polarization field interaction. The relation between the energy, T_e, and the wave vector, $\vec{k}$, of the electron may in general be quite complicated, but by restricting attention to an electron of energy near the bottom of the conduction band, the energy of the electron measured relative to the bottom of the conduction band can be presumed to be of the simple form

$$T_e = (\hbar\vec{k})^2/(2m) \tag{5}$$

in the case of isotropic solids, for which the Bloch mass m is a scalar. Also, for a sufficiently large crystal, $\vec{k}$ is essentially a continuous variable, so that the energy versus wave-vector relation

(5) is the same as for a free particle of mass m. It will be presumed henceforth that the ionic solid is isotropic and that T_e is small enough that (5) is applicable. In particular physical applications, this assumption should be checked for consistency.

The Bloch approximation is not adequate for the description of a conduction electron in an ionic lattice because, in the vicinity of the electron, the unshielded Coulomb field of the electron will significantly deform the ions and displace them from their lattice sites. Neglecting deformation for the moment, the electric field, $\Delta \vec{E}$, experienced by the electron, and not accounted for in the Bloch approximation, is equal to the total field of the displaced ions minus the static field which would result if the ions were to remain fixed at their lattice sites. Hence $\Delta \vec{E}$ is the electric field which would be produced if each lattice site were occupied by a dipole moment equal to the displacement of the ion from that site times the charge of the ion. For a slowly moving electron, the electron's wavelength will spread over many lattice sites, and for the purpose of calculating the effective value of $\Delta \vec{E}$ experienced by the electron, macroscopic dielectric methods may be employed. Accordingly (in the cgs Gaussian unit system)

$$\Delta \vec{E} = -4\pi \vec{P}, \tag{6}$$

where $\vec{P}$ is the macroscopic polarization field due to ionic displacement and is calculated as the average dipole moment per unit volume, averaged over a region which contains many lattice

sites, but which has a diameter small compared to the electron's wavelength. In a similar way, the Bloch approximation also neglects an electric field $\Delta \vec{E}_d$ experienced by the electron and arising from ionic deformation, with

$$\Delta \vec{E}_d = -4\pi \vec{P}_d \tag{7}$$

where $\vec{P}_d$ is the macroscopic polarization field due to ionic deformation.

The total macroscopic polarization field is

$$\vec{P}_T(\vec{r}) = \vec{P}(\vec{r}) + \vec{P}_d(\vec{r}), \tag{8}$$

and the total electric field experienced by the electron but neglected in the Bloch approximation is

$$\Delta \vec{E}_T = -4\pi \vec{P}_T . \tag{9}$$

The total macroscopic electric field in the solid is

$$\vec{E} = \vec{D} + \Delta \vec{E}_T , \tag{10}$$

where $\vec{D}$ is the electric displacement field, which is just the Coulomb field of the conduction electron, the only "free" charge present. Thus

$$\vec{D}(\vec{r}) = \frac{\partial}{\partial \vec{r}} \frac{e}{|\vec{r}-\vec{r}_e|} , \tag{11}$$

where $\vec{r}_e$ is the coordinate vector of the electron. In the absence of polarization effects, the ionic lattice produces no macroscopic electric field since it is macroscopically neutral.

It will be assumed henceforth that the electron's wave function does not significantly change over a region of many lattice sites so that the electric field $\Delta \vec{E}_T$ experienced by the Bloch

conduction electron is given by (9) wherein $\vec{P}_T$ is the total macroscopic electric polarization field (due to ionic deformation and displacement of ions from their lattice sites). Again, in particular applications, this assumption should be checked for consistency.

2. The Polarization Field[9]

One of the sources of the polarization field is the displacement of ions from their normal positions in the lattice. This ionic displacement is a dynamic effect, since it in turn is caused by the presence of the moving electron. Following the usual procedure in lattice dynamics, one may consider the motions of the individual ions as the result of collective motions (the normal modes) of the entire lattice. Each of these modes displaces the ions, producing dipole moments in the medium. The contribution of each mode to the polarization vector is determined by averaging the resultant dipole moments over a region of diameter large compared to the lattice spacing but small compared to the wavelength of the electron.

The normal modes may be divided roughly into two groups: the acoustical modes and the optical modes. In an acoustical mode, all the ions in one half-wavelength region between nodal planes vibrate in one direction, all those in the next half-wavelength region vibrate in the opposite direction, and so on. Over most of the lattice, therefore, adjacent ions vibrate in the same direction.

In an ionic crystal, adjacent ions are oppositely charged, so in an acousitcal mode, neighboring dipole moments are predominantly oppositely directed, and in the averaging process there is much cancellation. In an optical mode, adjacent ions in all half-wavelength regions between nodal planes vibrate in opposite directions; neighboring dipole moments are parallel and add constructively. The resulting contribution to the polarization is large compared to the contribution of an acoustical mode, and in comparison one is justified in entirely neglecting the polarization field due to the acoustical modes. Furthermore, the total dipole moments in each of two successive half-wavelength regions between the nodal planes of an optical mode are oppositely directed. As a result, destructive averaging occurs for short wavelengths, and constructive averaging occurs for long wavelengths. It is concluded, therefore, that the major contribution of ionic displacements to the polarization field comes from the long wavelength optical modes.

The polarization field due to ionic displacement, $\vec{P}(\vec{r})$, will be expressed in terms of a scalar potential $\phi(\vec{r})$ defined by

$$-4\pi \vec{P}(\vec{r}) = -\frac{\partial}{\partial \vec{r}} \phi(\vec{r}). \tag{12}$$

The potential energy of interaction of the Bloch conduction electron with the field is

$$V_1 = -e\phi(\vec{r}_e). \tag{13}$$

Using the particular form

$$\delta(\vec{r}-\vec{r}_e) = -\frac{1}{4\pi}\frac{\partial^2}{\partial r^2}\frac{1}{|\vec{r}-\vec{r}_e|} \tag{14}$$

for the Dirac δ-function, one finds that

$$\phi(\vec{r}_e) = \int \delta(\vec{r}-\vec{r}_e)\,\phi(\vec{r})\,d^3\vec{r} \tag{15}$$

$$= \int \vec{P}(\vec{r})\cdot\frac{\partial}{\partial\vec{r}}\frac{1}{|\vec{r}-\vec{r}_e|} \tag{16}$$

$$= \int \frac{\frac{\partial}{\partial\vec{r}}\cdot\vec{P}(\vec{r})}{|\vec{r}-\vec{r}_e|}\,d^3\vec{r}\ . \tag{17}$$

where obtaining the second and third of these equalities requires integration by parts. (The surface integrals which result can be made to vanish if the volume of integration is bounded by a surface entirely outside the material, where $\phi(\vec{r})$ and all its derivatives are zero). Use of (11), (13), and (16) yields

$$V_I = -\int \vec{P}(\vec{r})\cdot\vec{D}(\vec{r})\,d^3\vec{r}. \tag{18}$$

Since the divergence of a transverse wave is zero, (17) shows that the transverse modes of $\vec{P}(\vec{r})$ do not contribute to $\phi(\vec{r})$ and hence do not interact with the electron. Of the displacement

modes, therefore, only the long wavelength longitudinal optical modes interact significantly with the electron. These have a characteristic frequency ω_o which is virtually independent of wave vector.

The ionic deformations also have characteristic frequencies ω_d, where $\omega_d \gg \omega_o$. As the electron reaches a point $\vec{r}_e$ in its motion, the deformation polarization will tend toward the equilibrium configuration it would have if the electron were at rest at $\vec{r}_e$ much more rapidly than the displacement polarization, due to the relatively large value of ω_d. By symmetry, the closer a polarization field approaches its equilibrium configuration, the less force it exerts on the electron. Hence, the effect of the deformation polarization on a slowly moving electron will be neglected in comparison with the much greater force exerted by the displacement polarization. In terms of interaction with the electron, therefore, one is concerned only with the polarization due to the long wavelength longitudinal optical modes of lattice displacement, $\vec{P}(\vec{r})$, which have a single characteristic frequency of free oscillation, ω_o.

Consider the polarization of the ionic solid resulting from an applied electric displacement field $\vec{D}$. The total polarization $\vec{P}_T(\vec{r})$ can be resolved into

$$\vec{P}_T = \vec{P} + \vec{P}_d, \tag{19}$$

where $\vec{P}_d$ is due to ionic deformation. $\vec{P}_T$ satisfies the equation

$$\vec{E} = \vec{D} - 4\pi P_T , \tag{20}$$

where $\vec{E}$ is the total (macroscopic) electric field. In the case of static fields,

$$\vec{P}_T = \epsilon \vec{E}, \tag{21}$$

where ϵ is the static dielectric constant. Hence, for the static case,

$$\vec{P}_T = \frac{1}{4\pi}\left(1 - \frac{1}{\epsilon}\right)\vec{D}. \tag{22}$$

A similar equation involving $\vec{P}_d$ can be obtained if one applies to the medium an alternating displacement field of frequency ω' such that $\omega_d >> \omega' >> \omega_o$. The ionic displacement polarization $\vec{P}$ would then react so slowly as to respond only to the time averaged zero field, and hence $\vec{P}$ would vanish. The deformation oscillations would react so rapidly as to respond in essentially the same way that they would to a static field. Thus, for a static displacement field one obtains

$$\vec{P}_d = \frac{1}{4\pi}\left(1 - \frac{1}{\epsilon_o}\right)\vec{D}, \tag{23}$$

where ϵ_o is a dielectric constant characteristic of the medium at the frequency ω'. Combining (19), (22), and (23), one obtains, for the static case

$$\vec{P} = \frac{1}{4\pi}\left(\frac{1}{\epsilon_o} - \frac{1}{\epsilon}\right)\vec{D}. \tag{24}$$

Characteristically, ω_o is in the infrared region, ω' in the optical region, and ω_d in the ultraviolet absorption region. Hence ϵ_o

represents the optical dielectric constant before the onset of ultraviolet absorption. It may be written as $\epsilon_o = n^2$, where n is the optical refractive index of the medium.

3. The Classical Hamiltonian

The polarization field will be resolved into its normal modes in the form

$$\vec{P}(\vec{r}) = N\int \vec{A}(\vec{k})\, e^{i\vec{k}\cdot\vec{r}}\, d^3\vec{k} \tag{25}$$

$$\vec{A}(\vec{k}) = M\int \vec{P}(\vec{r})\, e^{-i\vec{k}\cdot\vec{r}}\, d^3\vec{r} \tag{26}$$

where N and M are constants. $\vec{A}(\vec{k})$ is the amplitude of the polarization field $\vec{P}(\vec{r})$ with wave vector $\vec{k}$ and must be proportional to the corresponding amplitude of the longitudinal optical lattice modes which produce $\vec{P}(\vec{r})$. Hence the kinetic energy T_p associated with the polarization field $\vec{P}$ is proportional to

$$\int \left|\dot{\vec{A}}(\vec{k})\right|^2 d^3\vec{k} \tag{27}$$

and may be expressed by use of (26) as

$$T_p = \int \tfrac{1}{2}\beta \left[\dot{\vec{P}}(\vec{r})\right]^2 d^3\vec{r}, \tag{28}$$

where β is a proportionality constant. The potential energy V_p

for the polarization field must be

$$V_p = \int \frac{1}{2} \beta \omega_o^2 \left[\vec{P}(\vec{r})\right]^2 d^3\vec{r} \tag{29}$$

in order for $\vec{P}$ to have a free oscillation frequency ω_o corresponding to the frequency of the long wavelength longitudinal optical modes which produce $\vec{P}$.

The Lagrangian for the system is constructed in the form

$$\begin{aligned} \mathcal{L} &= T_e + T_p - V_p - V_I \\ &= \frac{1}{2} m |\dot{\vec{r}}_e|^2 + \int \left\{ \frac{\beta}{2} \left[\dot{\vec{P}}(\vec{r})\right]^2 \right. \\ &\quad \left. - \frac{\beta}{2} \omega_o^2 \left[\vec{P}(\vec{r})\right]^2 + \vec{P}(\vec{r}) \cdot \vec{D}(\vec{r}) \right\} d^3\vec{r}. \end{aligned} \tag{30}$$

(31)

where V_I is taken from (18). The momenta conjugate to $\vec{r}_e$ and $\vec{P}(\vec{r})$, taken as generalized coordinates for the system, are $\vec{p}_e = m\dot{\vec{r}}_e$ and $\pi(\vec{r}) = \beta\dot{\vec{P}}(\vec{r})$ respectively. The Lagrangian equation of motion for the polarization field is

$$\ddot{\vec{P}}(\vec{r}) + \omega_o^2 \vec{P}(\vec{r}) = (1/\beta)\vec{D}(\vec{r}), \tag{32}$$

which must reduce to (24) in the steady state limit where the second order time derivative of $\vec{P}$ vanishes. Hence

$$\frac{1}{\beta} = \frac{\omega_o^2}{4\pi} \left(\frac{1}{\epsilon_o} - \frac{1}{\epsilon} \right). \tag{33}$$

The classical Hamiltonian H_c is then given by

$$H_c = \vec{P}_e \cdot \vec{F}_e + \int \vec{\Pi}(\vec{r}) \cdot \vec{P}(\vec{r})\, d^3\vec{r} - \mathcal{L} \tag{34}$$

$$= \frac{\vec{P}_e^{\,2}}{2m} + \int \left[\frac{\Pi^2(\vec{r})}{2\beta} + \frac{\beta}{2}\omega_o^2 P^2(\vec{r}) \right] d^3\vec{r} + e\phi(\vec{r}) \tag{35}$$

with the Poisson brackets

$$\left\{ (\vec{F}_e)_i , (\vec{P}_e)_j \right\} = \delta_{ij} \tag{36}$$

and

$$\left\{ P_i(\vec{r}), \Pi_j(\vec{r}') \right\} = \delta_{ij}\, \delta(\vec{r}-\vec{r}'). \tag{37}$$

The quantities $\vec{P}$, $\vec{\pi}$, and ϕ are now to be expanded into Fourier components preparatory to quantization of the polarization field. The set of functions

$$\left\{ V^{-\frac{1}{2}} (\vec{w}/w) e^{i\vec{w}\cdot\vec{r}} \;\middle|\; \vec{w} \right\} \tag{38}$$

forms a complete orthonormal set of vector functions (for expansions of longitudinal fields) subject to periodic boundary conditions over a cube of volume $V=L^3$, provided each component, w_i, of $\vec{w}$ satisfies

the condition

$$w_l = (2\pi/L)n_l \tag{39}$$

for some integer n_l. In the limit of large normalization volume V, $\vec{w}$ becomes a continuous variable, and sums may be converted to integrals by the replacement

$$\sum_{\vec{w}} \Longrightarrow \frac{V}{(2\pi)^3}\int d^3\vec{w}\,. \tag{40}$$

The (real valued) fields $\vec{P}(\vec{r})$ and $\vec{\pi}(\vec{r})$ may be expanded into the Fourier forms

$$\vec{P}(\vec{r}) = \left(\frac{\hbar}{2\beta\omega_0 V}\right)^{\frac{1}{2}} \sum_{\vec{w}} \frac{\vec{w}}{w}\left(b^*_{\vec{w}}\, e^{-i\vec{w}\cdot\vec{r}} + c.c.\right) \tag{41}$$

and

$$\vec{\pi}(\vec{r}) = i\left(\frac{\hbar\beta\omega_0}{2V}\right)^{\frac{1}{2}} \sum_{\vec{w}} \frac{\vec{w}}{w}\left(b^*_{\vec{w}}\, e^{-i\vec{w}\cdot\vec{r}} - c.c.\right). \tag{42}$$

By inspection and reference to (12), it can be seen that

$$\phi(\vec{r}_e) = 4\pi i\left(\frac{\hbar}{2\beta\omega_0 V}\right)^{\frac{1}{2}} \sum_{\vec{w}} \frac{1}{w}\left(b^*_{\vec{w}}\, e^{-i\vec{w}\cdot\vec{r}_e} - c.c.\right). \tag{43}$$

The substitution of these expansions into (35) yields

$$H_c = \frac{\vec{p}_e^{\,2}}{2m} + \hbar\omega_0 \sum_{\vec{w}} b^*_{\vec{w}}\, b_{\vec{w}}$$

$$+ 4\pi i\left(\frac{e^2\hbar}{2\beta\omega_0 V}\right) \sum_{\vec{w}} \frac{1}{w}\left(b^*_{\vec{w}}\, e^{-i\vec{w}\cdot\vec{r}_e} - c.c.\right). \tag{44}$$

Equations (41) and (42) can be inverted to obtain

$$b_{\vec{w}} = \left(\frac{\beta\omega_o}{2\hbar V}\right)^{\frac{1}{2}} \frac{\vec{w}}{w} \int \left[\vec{\rho}(\vec{r}) + \frac{i}{\beta\omega_o}\vec{\pi}(\vec{r})\right] e^{-i\vec{w}\cdot\vec{r}} d^3\vec{r}. \tag{45}$$

Equations (37) and (45), together with the definition of the Poisson bracket, can be used to calculate

$$\{b_{\vec{w}}, b_{\vec{w}'}\} = \{b_{\vec{w}}^*, b_{\vec{w}'}\} = 0 \tag{46}$$

and

$$\{b_{\vec{w}}, b_{\vec{w}'}^*\} = \frac{1}{i\hbar}\delta_{\vec{w},\vec{w}'}. \tag{47}$$

It is convenient at this point to introduce a natural system of units (equivalent in effect to setting $\hbar = \omega_o = 2m = 1$) in which length is measured in units of

$$u = [\hbar/(2m\omega_o)]^{\frac{1}{2}}, \tag{48}$$

time in units of ω_o^{-1}, and mass in units of 2m. This leads to the measurement of momentum in units of $\hbar/u$ and of energy in units of $\hbar\omega_o$. The following dimensionless quantities are introduced:

$$H = H_c/(\hbar\omega_o) \tag{49}$$

$$S = u^{-3}V \tag{50}$$

$$\vec{x} = u^{-1}\vec{r}_e \tag{51}$$

$$\vec{v} = u\vec{w} \tag{52}$$

$$\vec{K} = (u/\hbar)\vec{p}_e \tag{53}$$

$$\alpha = \frac{1}{2}\left(\frac{1}{n^2} - \frac{1}{\epsilon}\right)\frac{e^2}{\omega\hbar\omega_0} \tag{54}$$

$$\gamma_0 = (4\pi\alpha/S) \tag{55}$$

The introduction of these quantities into (44) produces the dimensionless Hamiltonian

$$H = \vec{K}^2 + \sum_{\vec{\nu}} b^*_{\vec{\nu}} b_{\vec{\nu}} + i\gamma_0 \sum_{\vec{\nu}} \frac{1}{\nu}\left(b^*_{\vec{\nu}}\, e^{-i\vec{\nu}\cdot\vec{x}} - c.c.\right). \tag{56}$$

Equation (40) may be rewritten in the form

$$\gamma_0^2 \sum_{\vec{\nu}} \Longrightarrow \frac{\alpha}{2\pi^2}\int d^3\vec{\nu} \tag{57}$$

and (36) in the form

$$\left\{ x_i, K_j \right\} = \frac{1}{\hbar}\,\delta_{i,j}\,. \tag{58}$$

Dimensionless forms of the other preceding equations will not be required. One should note that all Poisson brackets have been expressed with respect to the original (dimensional) coordinates and momenta and hence that the Poisson brackets of dimensionless quantities have units of reciprocal action.

4. The Quantum Mechanical Hamiltonian

The first requirement for quantization is that all products of real dynamical variables be written in symmetrical form. Thus (56) is written

$$H = K^2 + \frac{1}{2}\sum_{\vec{\nu}} \left(b_{\vec{\nu}}^{*} b_{\vec{\nu}} + b_{\vec{\nu}} b_{\vec{\nu}}^{*} \right) + i\gamma_0 \sum_{\vec{\nu}} \frac{1}{\nu} \left(b_{\vec{\nu}}^{*} e^{-i\vec{\nu}\cdot\vec{x}} - c.c. \right). \tag{59}$$

The fundamental dynamical variables $\vec{x}$, $\vec{K}$, $b_{\vec{\nu}}$, and $b_{\vec{\nu}}^{*}$ (in terms of which all other dynamical variables may be expressed) will be replaced by quantum mechanical operators obeying commutation rules given by the prescription

$$[O_1, O_2] = i\hbar \{O_1, O_2\}, \tag{60}$$

where O_1 and O_2 denote operators or corresponding dynamical variables as appropriate. Except for the replacement of * (denoting complex conjugation of a dynamical variable) by the more conventional $^{+}$ (denoting Hermitian conjugation of an operator), no explicit notation will be introduced to denote the new operator status of the old dynamical variables.

The following commutation rules follow from (46), (47), (58), and (60):

$$\left[b_{\vec{\nu}}, b_{\vec{\nu}'} \right] = \left[b_{\vec{\nu}}^{+}, b_{\vec{\nu}'}^{+} \right] = 0 \tag{61}$$

$$[b_{\vec{\nu}}, b_{\vec{\nu}'}^{+}] = \delta_{\vec{\nu},\vec{\nu}'} \tag{62}$$

$$[x_i, K_j] = i\,\delta_{i,j} \tag{63}$$

As is frequently convenient, (63) will be satisfied by taking the operator form of $\vec{x}$ as denoting multiplication by $\vec{x}$ and the operator form of $\vec{K}$ as denoting the differential operator

$$\vec{K} = \frac{1}{i}\frac{\partial}{\partial \vec{x}} . \tag{64}$$

By use of (61) and (62) one may simplify the second term in (59) so that

$$H = K^2 + \sum_{\vec{\nu}} b_{\vec{\nu}}^{+} b_{\vec{\nu}} + i\gamma_0 \sum_{\vec{\nu}} \frac{1}{\nu}\left(b_{\vec{\nu}}^{+} e^{-i\vec{\nu}\cdot\vec{x}} - h.c.\right), \tag{65}$$

where a constant term

$$\frac{1}{2}\sum_{\vec{\nu}} 1_{\vec{\nu}} \tag{66}$$

has been omitted. Unless one introduces a short wavelength cutoff for the wave vector $\vec{\nu}$, such a term is infinite; in any case, it is a constant representing the zero point energy of the longitudinal optical modes and does not affect the dynamics of the problem. The

three terms in (65) represent, in units of $\hbar\omega_0$, the following energies:

1. K^2 represents the kinetic energy of a free Bloch electron, measured from the bottom of the conduction band of the ionic solid.

2. The term

$$\sum_{\vec{v}} b^{+}_{\vec{v}} b_{\vec{v}} \tag{67}$$

represents the total energy of the longitudinal optical modes of oscillation of the ions in the absence of interaction with the conduction electron.

3. The remaining term represents the interaction energy of the Bloch conduction electron and the longitudinal optical modes of oscillation of the ions.

The commutation relations (61) and (62) indicate that $b^{+}_{\vec{v}}$ and $b_{\vec{v}}$ can be regarded as creation and destruction operators for a boson (a phonon corresponding to the longitudinal optical lattice modes) of wave vector $\vec{v}$; $b^{+}_{\vec{v}} b_{\vec{v}}$ is the operator for the number of phonons of wave vector $\vec{v}$.

The normalized eigenfunctions of the Hamiltonian (67) of the free phonon field will be written as

$$|n_{\vec{v}}\rangle, \tag{68}$$

which represents a state in which there are $n_{\vec{v}_1}$ phonons of wave vector $\vec{v}_1$, $n_{\vec{v}_2}$ phonons of wave vector $\vec{v}_2$, etc., where $\vec{v}_1$, $\vec{v}_2$... are all of the possible wave vectors defined by (39) and (52).

The shortened notation

$$\Phi_o \tag{69}$$

will be used for the phonon vacuum state.

5. Introduction of the Magnetic Field[10]

The Hamiltonian for a polaron in a constant uniform magnetic field will now be obtained. The magnetic field $\vec{B}$ will be directed along the z-axis of a rectangular cartesian coordinate system with coordinates x, y, and z. It will be described by the vector potential

$$A = (-By,0,0) \tag{70}$$

(known as the Landau gauge) and will be treated as a classical field. The medium will be presumed to be magnetically inert. The polaron Hamiltonian with the magnetic field present can be obtained from the field free polaron Hamiltonian by replaceing the electron's ordinary momentum $\vec{p}_e$ by

$$\vec{p}_e + (e/c)\vec{A}(\vec{r}_e), \tag{71}$$

where $\vec{p}_e$ then represents the electron's canonical momentum. In terms of the dimensionless quantities previously defined, this is equivalent to replacing the electron's dimensionless momentum $\vec{K}$ (defined in (53)) by

$$\vec{K} + (ue/\hbar c)\vec{A}(ux) \tag{72}$$

in the equation (65) for the dimensionless field free polaron Hamiltonian. Since, after this replacement, $\vec{K}$ is still the

dimensionless form of the electron's canonical momentum, it still obeys (63) and (64).

With the definitions

$$\omega_c = Be/(mc) \tag{73}$$

and

$$\omega = \omega_c/\omega_0 \quad , \tag{74}$$

the expression (72) may be written as

$$\vec{K}' \equiv \vec{K} + (-\tfrac{1}{2}\omega x_2, 0, 0). \tag{75}$$

This expression is to be substituted for $\vec{K}$ into (65) to obtain the Hamiltonian for the polaron in a uniform constant magnetic field described by (70). In summary, the result is

$$H = K'^2 + \sum_{\vec{v}} b^\dagger_{\vec{v}} b_{\vec{v}} + i\gamma_0 \sum_{\vec{v}} \frac{1}{v} (b^\dagger_{\vec{v}} e^{-i\vec{v}\cdot\vec{x}} - h.c.) \tag{76}$$

with:

$$\vec{K}' = \vec{K} + (-\tfrac{1}{2}\omega x_2, 0, 0) \tag{77}$$

$$\vec{K} = \frac{1}{i}\frac{\partial}{\partial \vec{x}} \tag{78}$$

$$\gamma_0 = (4\pi\alpha/S)^{\frac{1}{2}} \tag{79}$$

$$\omega_c = Be/(mc) \tag{80}$$

$$\omega = \omega_c/\omega_0 \tag{81}$$

$$\gamma_0^2 \sum_{\vec{\nu}} \Rightarrow \frac{\alpha}{2\pi^2} \int d^3\vec{\nu} \tag{82}$$

It is sometimes convenient to introduce the operator

$$a = -(1/\sqrt{\omega})(K_1 - \tfrac{1}{2}\omega x_2 - iK_2) \tag{83}$$

in terms of which

$$\vec{K}' = \frac{\sqrt{\omega}}{2}\left([a^+ + a], i[a^+ - a], \frac{2}{\sqrt{\omega}} K_3\right). \tag{84}$$

The operators a^+ and a commute with all the $b_{\vec{\nu}}^+$ and $b_{\vec{\nu}}$, and satisfy the commutation rule

$$[a, a^+] = 1 \tag{85}$$

for boson creation and destruction operators.

The term K'^2 alone is the Hamiltonian for the Bloch conduction electron in the magnetic field $\vec{B}$ without interaction with the lattice modes. The eigenvalue problem for this Hamiltonian can be solved exactly in the form

$$K'^2 \psi(n, k_1, k_3) = [(n + \tfrac{1}{2})\omega + k_3^2]\, \psi(n, k_1, k_3). \tag{86}$$

where k_1 and k_3 are real continuous quantum numbers ranging from $-\infty$ to $+\infty$ and where n is a non-negative integer quantum number. In terms of the Hermite polynomials $h_n(z)$,

$$\psi(n, k_1, k_3) = e^{i(k_1 x_1 + k_3 x_3) - \frac{\omega}{4}(x_2 - 2k_1/\omega)^2} h_n\left[\left(\frac{\omega}{2}\right)^{\frac{1}{2}}(x_2 - 2k_1/\omega)\right] \tag{87}$$

apart from normalization constants.

6. A Preliminary Transformation

It is sometimes convenient to represent the operators for real dynamical variables of interest in a new representation obtainable from the old one previously used by applying the unitary transformation

$$U_1 = e^{-i\vec{x}\cdot\vec{w}} \tag{88}$$

where

$$\vec{w} = \sum_{\vec{v}} \vec{v}\, b_{\vec{v}}^{+} b_{\vec{v}} . \tag{89}$$

One may obtain the following results:

Phonon destruction operators $= b_{\vec{v}} \Rightarrow e^{-i\vec{v}\cdot\vec{x}} b_{\vec{v}}$ (90)

Phonon creation operators $= b_{\vec{v}}^{+} \Rightarrow e^{i\vec{v}\cdot\vec{x}} b_{\vec{v}}^{+}$ (91)

Total phonon wave vector $= \vec{w} = \sum_{\vec{v}} \vec{v}\, b_{\vec{v}}^{+} b_{\vec{v}} \Rightarrow \vec{w}$ (92)

Electron coordinate vector $= \vec{x} \Rightarrow \vec{x}$ (93)

Electron canonical wave vector $= \vec{K} = \frac{1}{i}\frac{\partial}{\partial \vec{x}} \Rightarrow \vec{K} - \vec{w}$ (94)

Electron kinetic wave vector $= \vec{K}' = \vec{K} + (-\frac{1}{2}\omega x_2, 0, 0) \Rightarrow \vec{K}' - \vec{w}$ (95)

Total canonical wave vector $= \vec{K} + \vec{w} \Rightarrow \vec{K}$ (96)

Total Hamiltonian (76) $\Rightarrow$

$$H = (\vec{K}' - \vec{w})^2 + \sum_{\vec{v}} b_{\vec{v}}^{+} b_{\vec{v}} + i\gamma_0 \sum_{\vec{v}} (b_{\vec{v}}^{+} - b_{\vec{v}}) \tag{97}$$

with

$$\vec{K}' = \vec{K} + (-\tfrac{1}{2}\omega x_2, 0, 0)$$

$$\vec{K} = \frac{1}{L}\frac{\partial}{\partial \vec{x}}$$

$$\vec{w} = \sum_{\vec{v}} \vec{v}\, b^{+}_{\vec{v}} b_{\vec{v}} \tag{98}$$

where

$$\omega = \omega_c/\omega_o$$

$$\omega_c = Be/(mc) \tag{99}$$

and

$$\gamma_o^2 \sum_{\vec{v}} \Longrightarrow \frac{\alpha}{2\pi^2} \int d^3\vec{v} \; . \tag{100}$$

CHAPTER 3

THE FREE POLARON

1. Introduction

When no magnetic field is present, (97) is cyclic in $\vec{x}$. The total wave vector operator, $\vec{K}$, is then conserved and can be replaced by a c-number, $\vec{k}$, in (97) to produce the Hamiltonian operator

$$H(\vec{k}) = (\vec{k} - \vec{w})^2 + \sum_{\vec{v}} b_{\vec{v}}^{\dagger} b_{\vec{v}} + i\gamma_o \sum_{\vec{v}} \frac{1}{v} (b_{\vec{v}}^{\dagger} - b_{\vec{v}}) \tag{101}$$

the ground state of which describes a free (ground state) polaron of wave vector $\vec{k}$. For sufficiently small $\vec{k}$, the ground state eigenvalue, $E(\vec{k})$, of $H(\vec{k})$ can be expanded in the form

$$E(\vec{k}) = E_o + gk^2 + O(k^4). \tag{102}$$

The first term, E_o, represents (in units of $\hbar\omega_o$) the energy of a free polaron at rest and is called the self-energy of the polaron. (The self-energy is also the exact ground state eigenvalue of the free polaron Hamiltonian (65), whose ground state eigenfunction is simultaneously an eigenfunction of the total wave vector operator with eigenvalue zero). The second term of (102) represents (in units of $\hbar\omega_o$) the kinetic energy of a slowly moving polaron. The effective mass, m^*, of a free polaron at rest, called the polaron mass, may

then be defined by

$$(gk^2)\hbar\omega_o = p^2/(2m^*), \tag{103}$$

where

$$\vec{p} = (\hbar/u)\vec{k} \tag{104}$$

is the momentum of the polaron (in ordinary units). One may combine (102), (103), and (104) to obtain

$$E(\vec{k}) = E_o + (1/\mu)k^2 + O(k^4), \tag{105}$$

where

$$\mu = m^*/m \tag{106}$$

is called the polaron mass ratio. This definition of the polaron effective mass and the polaron mass ratio will be referred to as Frohlich's definition.[3]

Once one obtains an approximation for $E(\vec{k})$, for small $\vec{k}$, then (105) provides corresponding approximations for the polaron self-energy, E_o, and mass ratio, μ. Many such calculations have been published. In this chapter those publications will be reviewed which meet the following criteria:

1. The ideas or techniques are applicable in the presence of a magnetic field.

2. The results which are obtained for the polaron self-energy and the polaron mass ratio are to be compared to the results which will later be obtained for the corresponding quantities in the magnetic field case.

The Feynman-Schultz[4,5] calculation meets these criteria in a sense, but the definition of effective mass used by these

authors is not that presented above and has not been shown to be equivalent to it. The results they obtain are in agreement with those presented in Sections 3 (for weak coupling) and 5 (for strong coupling) of this chapter.

2. Perturbation Theory[3]

If the coupling constant, α, is sufficiently small, the interaction (last) term in (101) can be treated as a perturbation. The first order results are

$$E_o = -\alpha + O(\alpha^2) \tag{107}$$

and

$$\mu = 1 + \alpha/6 + O(\alpha^2) \tag{108}$$

with a second order correction of $-0.016\alpha^2 + O(\alpha^3)$ to E_o.

3. The Lee-Low-Pines Variational Method for Weak Coupling[3,11,12,13]

Variational methods have the advantage of providing an upper bound for the ground state energy of a system. In practice, one most often assumes a trial state function, ψ_t, as a function of one or more variational parameters which are then chosen to minimize

$$E(\vec{R}) = \frac{\langle \psi_t | H(\vec{R}) | \psi_t \rangle}{\langle \psi_t | \psi_t \rangle} \ . \tag{109}$$

A weak coupling variational approximation suggested by perturbation theory is to try

$$\psi_t = U_2 \Phi_o \ , \tag{110}$$

where Φ_o is the phonon vacuum state and U_2 is the unitary transformation given by

$$U_2 = e^{-i\sum_{\vec{v}} (f_{\vec{v}} b_{\vec{v}}^{+} + h.c.)}, \tag{111}$$

because, with a suitable choice of the function $f_{\vec{v}}$, U_2 transforms to zero all terms of $H(\vec{k})$ which are linear in phonon creation and destruction operators. The same choice of $f_{\vec{v}}$ is obtained by minimizing (109) with respect to $f_{\vec{v}}$ when (110) and (111) are employed. A state function of the form of (110) with (111) has the following interpretation: It is the most general form of a state function such that the probability of finding n_1 phonons of wave vector $\vec{v}_1$, n_2 phonons of wave vector $\vec{v}_2$, etc. is given by the simple product form

$$|f_{\vec{v}_1}|^{2n_1} |f_{\vec{v}_2}|^{2n_2} \cdots, \tag{112}$$

in which case the phonons in the field are said to be statictically uncorrelated. Statistical correlations are negligible for sufficiently weak coupling.

One finds that under U_2

$$b_{\vec{v}} \Rightarrow b_{\vec{v}} - i f_{\vec{v}}. \tag{113}$$

It is then easy to evaluate (109). The result is

$$E(\vec{k}) = \varepsilon^2 + \sum_{\vec{v}} (1+v^2) |f_{\vec{v}}|^2 - \gamma_o \sum_{\vec{v}} \frac{1}{v} (f_{\vec{v}}^{*} + f_{\vec{v}}), \tag{114}$$

where

$$\vec{\mathcal{I}} = \sum_{\vec{v}} \vec{v} \, |f_{\vec{v}}|^2 \tag{115}$$

and

$$\vec{z} = \vec{k} - \vec{t}. \tag{116}$$

Minimization of (114) with respect to $f_{\vec{v}}$ yields

$$f_{\vec{v}} = \frac{\gamma_0}{v} \frac{1}{1 + v^2 - 2\vec{v}\cdot\vec{z}} . \tag{117}$$

One notes by inspection that $\vec{t} = \vec{z} = 0$ for $\vec{k} = 0$ so that $\vec{z} = 0(\vec{k})$ and

$$f_{\vec{v}} = \frac{\gamma_0}{v} \frac{1}{1+v^2} \left[1 + \frac{2\vec{v}\cdot\vec{z}}{1+v^2} + \frac{4(\vec{v}\cdot\vec{z})^2}{(1+v^2)^2} \right] + O(k^3). \tag{118}$$

Use of (114), (115), and (116) yields

$$\vec{t} = (\alpha/6)\vec{z} + 0(k^3), \tag{119}$$

$$\vec{z} = (\alpha/6)(1+\alpha/6)^{-1}\vec{k} + 0(k^3), \tag{120}$$

and

$$E(\vec{k}) = -\alpha + (1+\alpha/6)^{-1}k^2 + 0(k^3), \tag{121}$$

so that the results for the self-energy and the polaron mass ratio are the same as for first order perturbation theory. The advantage of this result is that it is variational so that, in particular, the exact self-energy cannot exceed $-\alpha$. That is

$$E_o\Big|_{\text{exact}} \leq -\alpha. \tag{122}$$

4. The Landau-Pekar Variational Method for Strong Coupling[2,14]

For sufficiently large α the frequency ω_o of the polarization field is relatively small, other quantities being considered constant. The polarization field is then slow to react to the electron, which sees a deep and nearly static potential well. Such a situation can be described approximately by a "self trapped" polaron model state function of the form

$$\Psi_t = \Omega(\vec{x}-\vec{y})\,\Phi(\vec{y}), \tag{123}$$

where $\Omega(\vec{x}-\vec{y})$ is the probability amplitude that the electron will be located at $\vec{x}$ if the polarization potential is centered at some arbitrary point $\vec{y}$, and where $\Phi(\vec{y})$ describes a polarization field centered at $\vec{y}$. For the purpose of calculating the polaron self-energy by use of (123), the variationally best choice for $\Phi(\vec{y})$ may be found by minimizing

$$E_o = \frac{\langle \Psi_t | H | \Psi_t \rangle}{\langle \Psi_t | \Psi_t \rangle} \tag{124}$$

with respect to $\Phi(\vec{y})$, where H is given by (65). The result is

$$\Phi(\vec{y}) = U_2 \Phi_o , \tag{125}$$

where U_2 is given by (111) with

$$f_{\vec{v}}(\vec{y}) = \frac{\gamma_o}{V} e^{-i\vec{v}\cdot\vec{y}} \int e^{-i\vec{z}\cdot\vec{v}} \left|\Omega(\vec{z})\right|^2 d^3\vec{z}. \tag{126}$$

Substituting (126) into (124) yields

$$E_o = \int \Omega^*(\vec{z}) \left(\frac{1}{i} \frac{\partial}{\partial \vec{x}} \right)^2 \Omega(\vec{z}) + \sum_{\vec{\nu}} |f_{\vec{\nu}}|^2 . \tag{127}$$

Minimization of this result with respect to $\Omega(\vec{z})$ is not practical. Instead, various trial forms of $\Omega(\vec{z})$, involving variational parameters, have been used, all with very similar results for the self-energy, E_o. As a convenient example, which will be useful later, the choice

$$\Omega(\vec{z}) = \left(\frac{2}{\pi \beta^2} \right)^{3/4} e^{-z^2/\beta^2} \tag{128}$$

yields

$$f_{\vec{\nu}}(\vec{y}) = \frac{\gamma_o}{\nu} e^{-\nu^2 \beta^2 / 8 - i \vec{\nu} \cdot \vec{y}} , \tag{129}$$

$$\beta = (3/\alpha)(2\pi)^{1/2} , \tag{130}$$

and

$$E_o = -\alpha^2/(3\pi) . \tag{131}$$

For values of the coupling constant, α, greater than about ten, this (variational)result for E_o is lower than, and therefore superior to, the result of the previous section. Moreover, that the self-energy should go as α^2 for strong coupling (large α) is apparently correct. Lieb and Yamazaki[15] have determined that, for asymtotically large α, the self-energy, E_o, has the lower bound $-(1/3)\alpha^2$. Thus, for sufficiently large α,

$$-\alpha^2/3 < E_o\Big|_{exact} < -\alpha^2/(3\pi). \qquad (132)$$

So, assuming that E_o goes as some power, p, of α as α goes to infinity, it is then known that p = 2.

Unfortunately, a trial state function of the self-trapped polaron form (123) is deficient in the respect that it is not an eigenfunction of the total wave vector and, as a result, is not suitable for calculating the polaron effective mass as defined by Frohlich. Frohlich's definition requires that the polaron energy be known as a function of its (conserved) wave vector. Landau and Pekar do calculate a polaron effective mass corresponding to the self-trapped polaron picture. They obtain a polaron mass ratio

$$\mu' = 1 + \left(\frac{2\alpha}{3\sqrt{\pi}}\right)^4 . \qquad (133)$$

However, their calculation is based on a reformulated definition of the polaron mass ratio, μ'. There is no demanding reason that the Landau-Pekar effective mass, as based on the self-trapped polaron picture, should approximate the Frohlich polaron mass.

5. The Hohler-Marshall Variational Method[18,19]

The preceding strong coupling approximation is based upon a trial state function representing a polaron trapped at a point $\vec{y}$ and having the form

$$\xi(\vec{x}-\vec{y},\vec{y}) = \Omega(|\vec{x}-\vec{y}|)U_2(\vec{y})\,\Phi_o \qquad (134)$$

with

$$U_2(\vec{y}) = e^{-i\sum_{\vec{v}}(f_{\vec{v}}(\vec{y})b_{\vec{v}}^{+} + h.c.)} \qquad (135)$$

where Ω and $f_{\vec{v}}$ are given in terms of the variational parameter β by (128) and (129) respectively. For the purpose of calculating the polaron mass, the difficulty with (134) is that it is not an eigenfunction of the total wave vector operator, $\vec{K} + \vec{w}$. There do exist, however, linear superpositions of states of the form (134) which are eigenfunctions of $\vec{K} + \vec{w}$. It will now be shown that the trial state function

$$\Psi_t = \int d^3\vec{y}\, e^{i\vec{R}\cdot(t\vec{y}+(1-t)\vec{x})}\, \xi(\vec{x}-\vec{y}, \vec{y}) \tag{136}$$

is such an eigenfunction for any value of the variational parameter t. First note that

$$U_1^{+}\Phi_o = \Phi_o, \tag{137}$$

where U_1 is given by (88). One can show, using (137), (90), (91), (134), and (135), that

$$U_1^{+}\xi(\vec{x}-\vec{y},\vec{y}) = \xi(\vec{x}-\vec{y},\vec{y}-\vec{x}). \tag{138}$$

Hence, with the aid of (78) and (96), one obtains

$$(\vec{K}+\vec{w})\Psi_t = U_1 U_1^{+}(\vec{K}+\vec{w})U_1 U_1^{+}\Psi_t \tag{139}$$

$$= U_1 \vec{K} U_1^{+}\Psi_t \tag{140}$$

$$= U_1 \vec{K} e^{i\vec{k}\cdot\vec{x}} \int d^3\vec{z}\, e^{it\vec{k}\cdot\vec{z}}\, \xi(-\vec{z},\vec{z}) \tag{141}$$

$$= \vec{k}\Psi_t. \tag{142}$$

Therefore Ψ_t is an eigenfunction of the total wave vector operator, $\vec{K} + \vec{w}$, with eigenvalue $\vec{k}$.

The variational parameters t and β are determined from minimizing

$$E(\vec{k}) = \frac{\langle \psi_t | H | \psi_t \rangle}{\langle \psi_t | \psi_t \rangle} , \tag{143}$$

where H is taken from (65).

The trial state function ψ_t was used by Hohler (with t = 1) to calculate E_o and μ. Improved results were obtained by Marshall, who introduced the additional variational parameter t. (The mass ratio obtained by Marshall is, in the limit of strong coupling, exactly twice that obtained by Hohler.) For t = 1, a trial state function of the form (136) may be interpreted as follows. The factor $\xi(\vec{x}-\vec{y},\vec{y})$ is the Landau-Pekar strong coupling wave function for a self-trapped polaron centered at a point $\vec{y}$, wherein the electron coordinate vector is $\vec{x}$. The factor $\exp(i\vec{k}\cdot\vec{y})$ is the probability amplitude that the polaron center will be found at $\vec{y}$, provided the polaron center behaves as a free particle. The calculations are long and complicated but involve only commonly known procedures. One finds that

$$E(\vec{k}) = E_o + \mu_{HM}^{-1} k^2 + O(k^4) , \tag{144}$$

where

$$E_o = \frac{\int R(x) Q(x) dx}{\int R(x) dx} , \tag{145}$$

$$\mu_{HM}^{-1} = 1 - \frac{\frac{2}{3}\left[\int x\,\ell(x)\,R(x)\,dx\right]^2}{\left[\int x^2 R(x)\,dx\right]\left[\int Q(x)R(x)\,dx\right] - \left[\int x^2 R(x)Q(x)\,dx\right]\left[\int R(x)\,dx\right]},$$

(146)

$$R(x) = x^2 e^{-x^2/2 + \frac{\alpha c}{2x}\phi(x)},$$

(147)

$$Q(x) = -\frac{2\alpha c}{x}\phi\left(\frac{x}{2}\right) + \frac{\alpha c}{4x}\left[2\phi(x) + c^2 x\,\phi'(x)\right].$$

(148)

$$\ell(x) = \frac{\alpha c^2}{4x^2}\left[x\,\phi'(x) - \phi(x)\right].$$

(149)

$$\phi'(x) = 2\pi^{-1/2} e^{-x^2},$$

(150)

$$\phi(x) = \int_0^x \phi'(y)\,d\vec{y},$$

(151)

and where the variational parameter, β has been replaced by

$$\beta = 2/c. \qquad (152)$$

There still remain one dimensional integrals which require numerical evaluation, for each value of the coupling constant, α, in order

that one may determine the remaining variational parameter c (defined in (152)),as well as the self-energy E_o, and the effective mass ratio, μ. The results for various values of the coupling constant are given in the following table.

Coupling constant α	Variational parameter c	Self energy E_o	Mass ratio μ
1	1.96±.01	.776	1.21±.00
2	2.02±.01	1.59	1.53±.00
4	2.22±.01	3.40	2.89±.02
6	2.59±.01	5.59	8.05±.08
7	2.87±.01	6.92	16.2 ±.2
8	3.18±.01	8.46	33.9 ±.4
9	3.54±.01	10.2	67.4 ±.7
10	3.90±.01	12.2	121 ±1
11	4.26±.01	14.4	198 ±2
13	4.99±.01	19.5	440 ±3
15	5.73±.01	25.4	840 ±5
20	7.49±.01	44.0	2900 ±10
30	11.3 ±.1	97.0	15400±400

The errors quoted for μ arise from the stated error to which the optimum values of the variational parameter c were determined. To within the accuracy implied by the table, no error in E_o results from this source. These results are in good agreement with previous calculations for all values of α (although the Lee-Low-Pines weak coupling results are somewhat superior for $\alpha \leq 7$).

The weak and strong coupling limits for the self-energy and polaron mass ratio are respectively

$$E_o = -0.778\alpha + O(\alpha^2) \tag{153}$$

$$\mu = 1 + 0.179\alpha + O(\alpha^2) \tag{154}$$

and

$$E_o = -(3\pi)^{-1}\alpha^2 - 3/2 + O(\alpha^{-2}) \tag{155}$$

$$\mu = (2\alpha/3\pi^{\frac{1}{2}}))^4 + O(\alpha^2). \tag{156}$$

In particular, (156) agrees, to highest order in α, with the Landau-Pekar result (133). Here, however, the result has been obtained using Frohlich's definition of effective mass and the improved trial state function (136). Unfortunately, there is still no demanding reason that the resultant Landau-Pekar forth power asymptotic dependence of the polaron mass ratio upon α should necessarily be correct.

CHAPTER 4

THE POLARON IN A MAGNETIC FIELD

1. Introduction

In the presence of a magnetic field the total wave vector is not conserved. The definitions of self-energy and effective mass ratio which were presented in the previous chapter are therefore not applicable. By inspection of (94) through (97), the $\vec{x}$ and $\vec{z}$ components of the total (canonical) wave vector are still conserved and may be set to zero by restricting further consideration to a description of polarons whose (canonical) wave vector is in a direction (taken to be the $\vec{y}$ direction) perpendicular to the magnetic field. With this restriction, for further reference, the Hamiltonian (97) is rewritten here as

$$H = (\vec{K}' - \vec{w})^2 + \sum_{\vec{v}} b_{\vec{v}}^{+} b_{\vec{v}} + i\gamma_0 \sum_{\vec{v}} \frac{1}{v} (b_{\vec{v}}^{+} - b_{\vec{v}}) \tag{157}$$

where

$$\vec{w} = \sum_{\vec{v}} \vec{v}\, b_{\vec{v}}^{+} b_{\vec{v}} , \tag{158}$$

$$\vec{K}' = (-\tfrac{1}{2}\omega x_2 , \tfrac{1}{i}\tfrac{\partial}{\partial x_2} , 0), \tag{159}$$

$$\omega = \frac{Be}{mc}\frac{1}{\omega_o} , \tag{160}$$

$$\gamma_o = (4\pi\alpha/S)^{\frac{1}{2}} , \tag{161}$$

and where, in the limit as the (unitless) normalization volume, S, approaches infinity,

$$\gamma_o^2 \sum_{\vec{v}} \Rightarrow \frac{\alpha}{2\pi^2}\int d^3\vec{v} \tag{162}$$

In the appendix it is shown that for a weak magnetic field (that is, ω small) the low-lying eigenvalues of (157) are given exactly by

$$E_n = E_o + (1/\mu)\ (n+\tfrac{1}{2})\omega + 0(\omega^2) \tag{163}$$

regardless of the value of the polaron coupling constant. In this result, $n = 0,1,2,\ldots$ and E_o and μ are respectively the polaron self-energy and mass ratio as defined by Frohlich for the free (ground state) polaron at rest.

A significant implication of (163) is that for a fixed ionic solid, in the limit of weak magnetic field, cyclotron resonant absorption will occur in the process of exciting ground state polarons (n=0) to the first level of magnetic excitation (n=1). For such a

process, the absorption frequency, ω_r, may be found (neglecting polaron recoil energy) by equating the energy, $\hbar\omega_r$, of an absorbed photon to the excitation energy $(\Delta\epsilon)\hbar\omega_o$ (in ordinary units) where, from (163),

$$\Delta\epsilon = \epsilon_1 - \epsilon_o = \omega/\mu \ . \tag{164}$$

Using (160) and (164), one obtains

$$\omega_r = eB/(m^*c), \tag{165}$$

where

$$m^* = \mu m \tag{166}$$

is Frohlich's polaron effective mass.

The result (163) is potentially useful for the purpose of calculating the polaron self-energy and mass ratio since in principle it allows one to determine these two quantities by approximating the ground state eigenvalue, ϵ_o, of (157) to first order in ω. In practice, one may not be certain a priori that his method of approximating the description of a polaron in a magnetic field will preserve the form of the exact result (163). In such an approximation it may be advisable for one to calculate his approximations for at lease ϵ_o and ϵ_1 to check his results for consistency with the form of (163) and to check that he obtains the rather accurately known results for E_o.

2. The Weak Coupling Case[17]

If the interaction (last) term in (157) is treated as a perturbation, to first order in α the results for the ground state and the first excited state are

$$\epsilon_o = -\alpha + (\omega/2)(1-\alpha/6) + 2\,\omega^2/240 + O(\omega^3) \qquad (167)$$

and

$$\epsilon_1 = -\alpha + (3/2)(1-\alpha/6)\omega - 7\alpha\omega^2/48 + O(\omega^3). \qquad (168)$$

Equation (163) then yields the expected results

$$E_o = -\alpha + O(\alpha^2) \qquad (169)$$

and

$$\mu = 1 + \alpha/6 + O(\alpha^2) \qquad (170)$$

in accord with what was found in the previous chapter.

By an extension of the Lee-Low-Pines weak coupling method discussed in the previous chapter, Larsen has calculated a low energy spectrum of (157). His results are consistent with (163), (169), and (170).

3. The Strong Coupling Case

In this section an attempt will be made to construct approximate state functions for the two lowest energy eigenstates of a strongly coupled polaron in a weak magnetic field. The trial state functions to be employed are constructed as follows, by analogy with Hohler's state function for a strongly coupled free polaron:

$$\int d^3\vec{y}\; g(\vec{y})\,\xi(\vec{x}-\vec{y},\vec{y}) \qquad (171)$$

where $\xi(\vec{x}-\vec{y},\vec{y})$ is the Landau-Pekar strong coupling probability amplitude for a polaron self trapped at a center point, $\vec{y}$, and where $g(\vec{y})$ is the probability amplitude of finding the polaron

center at a point $\vec{y}$. The factor ξ is given by (134), (135), (128), and (129) in terms of the variational parameter β. Whereas, in the field-free case, Hohler takes $g(\vec{y})$ to be of the form for the wave function of a free particle, in the present case $g(\vec{y})$ is taken to be of the form of the wave function for a particle in the magnetic field:

$$g(\vec{y}) = \begin{cases} e^{-\eta\omega y_2^2} & \text{for the ground state} \\ y_2\, e^{-\eta\omega y_2^2} & \text{for the first excited state} \end{cases} \tag{172}$$

where ω, given by (98), is proportional to the magnetic field, and η is a variational parameter.

It is convenient to consider the trial state function (171) after it has been transformed by the unitary transformation U_1 (defined in (88)). Use of (137) and (138) reveals that the transformed trial state function is

$$\int d^3\vec{z}\; g(\vec{z}+\vec{x})\,\xi(-\vec{z},\vec{z}). \tag{173}$$

(This form is appropriate for use with the Hamiltonian, transformed under U_1, given by (157)). One finds that the trial state function (173), while it yields the proper spectrum and the correct value of E_o, does not yield the expected result for μ. A refinement of (173) to the form

$$\Psi_t = \int d^3\vec{z}\; g(\vec{z}+t\vec{x})\, e^{-i\sigma\omega x_2 z_1}\,\xi(-\vec{z},\vec{z}) \tag{174}$$

where t and σ are additional variational parameters, will yield a variationally superior result and recover the Landau-Pekar mass ratio in the limit of strong coupling.

The variational parameters β, η, σ, and t are determined by minimizing

$$E = \frac{\langle \psi_t | H | \psi_t \rangle}{\langle \psi_t | \psi_t \rangle} \, . \tag{175}$$

where ψ_t is taken from (174) and H from (157). The minimization is carried out separately for the ground state and the first excited state. The actual calculation is rather complicated, but it involves only commonly known precedures. The four variational parameters turn out to be the same for the first excited state as for the ground state. Moreover, for the optimum values of the variational parameters, the trial state function for the excited state is orthogonal to the trial state function for the ground state (as required by the variational principle). The final result is

$$E = E_o + \tfrac{1}{2}\delta\omega\mu_{HM} \, , \tag{176}$$

where

$$\delta = \begin{cases} 1 \text{ for the ground state} \\ 3 \text{ for the first excited state} \end{cases} , \tag{177}$$

and where E_o and μ_{HM} are given respectively by (145) and (146). The result (176) exhibits the expected form (163) for the spectrum and yields the proper value of the self-energy, E_o. The calculated value of the mass ratio is exactly

$$\mu = \mu_{HM} \, . \tag{178}$$

CHAPTER 5

CONCLUSIONS

1. Summary

A polaron is a composite particle which presumably is produced when a charged particle is placed in a polarizable medium. The polaron consists of the charged particle and its self-produced polarization field. Following Frohlich, a Hamiltonian is developed to describe the polaron which results when an electron is placed in the otherwise vacant conduction band of an ionic solid. In dimensionless form, Frohlich's Hamiltonian is a function of a single parameter, α, called the polaron coupling constant, whose value depends upon the properties of the ionic solid. Although the properties of a conduction electron in an ionic solid are of intrinsic interest, it is hoped that investigation of the Frohlich Hamiltonian may result in methods which are also applicable to other similar problems of interest. For this reason, the polaron coupling constant, α, is regarded as an arbitrary positive parameter in terms of which the properties of the polaron are to be expressed. The Frohlich Hamiltonian is then modified, by conventional methods, to include the presence of a uniform constant magnetic field.

Frohlich's definition of the polaron self-energy, E_0, and mass ratio, μ, are then developed. These definitions depend upon

the fact that, when no magnetic field is present (and only then), the polaron's total wave vector is conserved. It is shown in the appendix, however, that the (ground state) polaron in a weak magnetic field exhibits the same energy spectrum as would be obtained for a point particle whose self-energy is equal to the Frohlich self-energy and whose mass is equal to the Frohlich polaron effective mass.

Various methods have been used to evaluate E_o and μ when no magnetic field is present, and several of these methods are discussed next. For weak coupling, the results $E_o = -\alpha$ and $\mu = 1 + \alpha/6$ are found, from perturbation theory, to be exact to first order in α. Similar results are obtained from a variational calculation due to Lee, Low, and Pines. For sufficiently strong coupling, it is found that E_o becomes exactly proportional to $-\alpha^2$. The correct asymptotic dependence of μ upon α is not reliably known. A variational method developed by Hohler and improved by Marshall is found to give acceptable results for E_o and to yield a value of μ consistent with other strong coupling theories (all of which use definitions of the polaron effective mass which have not been shown to be equivalent to Frohlich's definition[3] through Frohlich's polaron ~~and the Frohlich effective~~ mass is the one which would be measured in a cyclotron resonance experiment).

The problem of determining E_o and μ is next approached for the polaron in a magnetic field, considering, in particular, methods which construct explicit approximations to the magnetic spectrum of the ground state polaron. The expected weak coupling results may be obtained from perturbation theory or from a modification of the

Lee-Low-Pines method which is due to Larsen. A strong coupling calculation, analogous to the Hohler-Marshall calculation for the field-free polaron, is presented next. The proper form for the magnetic spectrum is obtained, and the expected value of E_o is found. The value obtained for μ agrees with the result obtained by Marshall.

2. Experimental Comparisons

Experimental comparisons would provide the most direct check on the validity of polaron theories. Unfortunately, experimental data is severely limited. There have been no measurements of the polaron self-energy, and there is no data at all for any materials with large values of the coupling constant.

In order for polaron theory to make any numerical predictions at all, the numerical value of the polaron coupling constant, α, is required. To determine α, by use of its definition (54), one needs to know the optical refractive index, n, the static dielectric constant, ϵ, the frequency of longitudinal optical modes, ω_o, and the Bloch mass, m. The first three of these quantities are known for many ionic solids of interest. But the Bloch mass, although useful for theoretical purposes, is fictional in the sense that one cannot "turn off" the interaction of the electron with the polarization field. Any comparison of polaron theory to experiment, therefore, can only be a consistency check between two independent measurements. For example, one might first measure polaron mobility in a particular medium and then calculate a value of α by employing

a theory which expresses the mobility in terms of the coupling constant (assuming the other quantities required to obtain α are known). The calculated value of α might then be used to predict the result of a cyclotron resonance experiment, which result could then be checked by performing the experiment.

Polaron mobility measurements and cyclotron frequency measurements are the only two independent experiments which have been performed on the polaron. Mobility values are available for many of the alkali halides,[6] all of which are found to have small coupling constants. Cyclotron resonance has been observed for some of these materials.[6] The following list gives some relevant numerical values for AgBr, which will serve an an example:[20,21]

$$\omega_o = 2.5 \times 10^{13} \text{sec}^{-1}$$
$$\epsilon = 13.1$$
$$n^2 = 4.6 \tag{179}$$

From (179) and (54), one obtains

$$\alpha = 4.0(m/m_e)^{\frac{1}{2}}. \tag{180}$$

Mobility in AgBr has been studied extensively as a function of temperature,[20] and, using (180), one may estimate

$$m = 0.23\ m_e$$
$$\alpha = 1.9$$
$$\mu m/m_e = 0.31 \tag{181}$$

by comparing the experimental results with the theory of Low and Pines.[22]

Brown and Ascarelli[6] have observed cyclotron resonance in AgBr at a frequency

$$\omega_r = 4.4 \times 10^{11} \text{sec}^{-1} \tag{182}$$

and a magnetic field

$$B = 6650 \text{ gauss}. \tag{183}$$

Since

$$\omega_r = eB/(\mu mc), \tag{184}$$

one finds that

$$\mu m/m_e = 0.27 \pm .01 , \tag{185}$$

where the estimated errors are reported by Brown and Ascarelli. If one assumes the weak coupling theoretical result $\mu = 1 + \alpha/6$, (180) and (185) lead to:

$$\alpha = 1.82$$

$$m = 0.21 m_e \tag{186}$$

(The parameter ω is defined in (98) and is used in the expansions in Chapter 4. The validity of the expansions requires that ω be small. From (179), (183), and (186) one may evaluate ω for the experiment of Brown and Ascarelli. One finds $\omega \approx 0.02$.)

The results (185) and (186) are in reasonable agreement with (181). If one accepts the values of the constants n and ϵ quoted in (179), agreement with other mobility theories is not nearly so good. Reported values of these constants vary, however, and better agreement with other theories is obtained if the values of n and ϵ are changed only slightly.

One can conclude only that, at present, experimental results are inadequate to distinguish different polaron theories.

3. Concluding Remarks

There are challenging reasons for finding the low-lying energy eigenstates for a strongly coupled polaron in a magnetic field as was attempted in Chapter 4. In the first place, it has never been done before. There are stringent checks by which one may judge such trial eigenstates for the case of weak magnetic fields; namely that the low-lying spectrum must have the form of the Frohlich polaron self-energy plus the spectrum for a point particle of the Frohlich polaron mass in the magnetic field. The polaron self-energy, E_o, is fairly reliably known for all values of the polaron coupling constant, α. The polaron mass ratio, μ, is not reliably known except for weak coupling, although a consensus of various approximate results would indicate that μ should be asymptotically proportional to α^4. A calculation of the low-lying spectrum for a polaron in a weak magnetic field therefore provides a new and independent approach to the problem of finding the polaron mass ratio.

There are two publications in which effective masses of strongly coupled polarons in weak magnetic fields are calculated (without attempting to obtain the low-lying energy states themselves). In each case the effective mass calculated is based on a definition peculiar to the approximation employed without proof that the definition used is equivalent to the Frohlich effective mass (which is what would be measured in a cyclotron resonance experiment). One of these is Tulub's calculation[23] which yields a mass ratio of $1 + \alpha/6$ for all α, in disagreement with all other strong coupling results.

The other is due to Hellwarth and Platzman[24], who use an extended version of the field free variational path integral method of Feynman and Schultz. Their results agree with the Feynman-Schultz effective mass, which is numerically approximated by the result of Marshall (given in the table on page 39).

For the purpose of calculating the low-lying energy spectrum of a strongly coupled polaron in a weak magnetic field, the only method which has been successful for a free polaron and which seems to lend itself to extension to the case of a polaron in a magnetic field is the Hohler-type approach. This method allows one to construct state functions based on the intuitive concept of a composite particle with fixed structure, the motion of whose center is appropriate for a point particle moving in the magnetic field. The resultant trial state functions employed in Chapter 4 display many of the properties which the exact low-lying energy eigenfunctions must possess. They are exact eigenfunctions of the conserved components of the polaron's wave vector. (This correct aspect is particularly significant in light of the fact that the only previous treatment which is consistent with the law of conservation of wave vector and which yields the Landau-Pekar asymptotic dependence of μ upon α is Marshall's Hohler-type calculation.) They also yield the exactly correct form for the dependence of the low-lying spectrum upon the magnetic field strength. (This achievement is not trivial since the trial ground state and excited state wave functions are properly orthogonal, in spite of their complexity, and both yield

the same values of E_o and μ.)

The result obtained for E_o comes out to be what one knows it should be. The result for μ agrees with the consensus of previous work. In view of the novelty of the approach taken and in view of the fact that the trial state functions used do display all of the proper features just discussed, this further agreement of the effective mass calculated provides a significant consistency with the bulk of research on the polaron problem.

APPENDIX

It is the purpose of this appendix to show that for a weak magnetic field (ω sufficiently small), the lowest-lying eigenvalues of the Hamiltonian (157), given by

$$H = H_o + K'^2 - 2\vec{K}'\cdot\vec{w} \tag{A1}$$

with

$$H_o = w^2 + \sum_{\vec{v}} b^+_{\vec{v}} b_{\vec{v}} + i\gamma_o \sum_{\vec{v}} \frac{1}{v}\left(b^+_{\vec{v}} - b_{\vec{v}}\right), \tag{A2}$$

$$\vec{w} = \sum_{\vec{v}} \vec{v}\, b^+_{\vec{v}} b_{\vec{v}}\,, \tag{A3}$$

$$\vec{K}' = \left(-\tfrac{1}{2}\omega x_2\,,\ \tfrac{1}{i}\frac{\partial}{\partial x_2}\,,\ 0\right), \tag{A4}$$

and

$$\gamma_o^2 \sum_{\vec{v}} \Rightarrow \frac{\alpha}{2\pi^2}\int d^3\vec{v}\,. \tag{A5}$$

have the form

$$\epsilon_n = E_o + \mu^{-1}(n+\tfrac{1}{2})\omega + O(\omega^2) \tag{A6}$$

where $n = 0,1,2,\ldots$ and where E_o and μ are the self-energy and

mass ratio of a (ground state) polaron at rest. The parameters E_o and μ are defined by

$$E_o(\vec{k}) = E_o + \mu^{-1}k^2 + O(k^2), \tag{A7}$$

where $E_o(\vec{k})$ is the ground state energy of the Hamiltonian for a polaron of wave vector $\vec{k}$, given by

$$H = H_o + k^2 - 2\vec{k}\cdot\vec{w}. \tag{A8}$$

The proof of (A6) is to be made independently of the magnitude of the polaron coupling constant, α.

The eigenstates and eigenvalues of H_o, defined by

$$H_o|E\rangle = E|E\rangle , \tag{A9}$$

are not known. It will be assumed, however, that the ground state $|E_o\rangle$ of H_o is non-degenerate and that the excited states of H_o have eigenenergies separated from its ground state energy, E_o, by a finite minimum amount of ΔE. Since H_o is invariant under rotation of spatial coordinates and E_o is non-degenerate, it follows that

$$\langle E_o|\vec{V}|E_o\rangle = 0 , \tag{A10}$$

$$\langle E_o|T_{ij}|E_o\rangle = 0 \text{ for } i\neq j , \tag{A11}$$

and

$$\langle E_o|T_{11}|E_o\rangle = \langle E_o|T_{22}|E_o\rangle = \langle E_o|T_{33}|E_o\rangle , \tag{A12}$$

where $\vec{V}$ is any vector and T_{ij} is any second rank tensor with respect to rotation of spatial coordinates.

If Schrodinger perturbation theory is employed, treating the last term of (A8) as perturbative, the result may be simplified

by use of (A10), (A11), and (A12) to yield (A7) with

$$\frac{1}{\mu} = 1 - 4 \sum_{|E\rangle}{}' \frac{|\langle E|W_3|E_o\rangle|^2}{E - E_o} \, .$$

(A13)

where the summation is over all excited states of H_o.

Perturbation theory may be similarly employed to treat the last term of (A1) as a perturbation to obtain the eigenvalues of (A1) as a power series in ω. In particular, to obtain the lowest lying eigenvalues of (A1), for small ω, one needs only to find the perturbation correction to the lowest lying eigenvalues, $\epsilon_n^{(0)}$, of $H_o + K'^2$. Since the excited states of H_o have energy greater than E_o by at least an amount ΔE, independent of ω, and since the eigenvalues of K'^2 (a simple harmonic oscillator Hamiltonian) are $\omega(n + \frac{1}{2})$ with $n = 0,1,2,\ldots$, then

$$\epsilon_n^{(0)} = E_o + \omega(n + \tfrac{1}{2}) \qquad \text{(A14)}$$

where E_o is the ground state eigenvalue of H_o, the self-energy of the polaron. (There are, of course, other states of $H_o + K'^2$ of higher energy corresponding to excited eigenstates of H_o, but for sufficiently small ω, there are many eigenstates of lower energy.) The result of applying perturbation theory and (A10), (A11), and (A12) to obtain the perturbation correction to (A14) is

$$\epsilon_n = \epsilon_n^{(0)} + 2\omega \sum_{|E\rangle}{}' |\langle E|W_3|E_o\rangle|^2 \left[\frac{n+1}{E_o - E - \omega} + \frac{n}{E_o - E + \omega} \right] + O(\omega^2) \, .$$

(A15)

Since $E - E_o \geq \Delta E$, the energy denominators occuring in (A15) do not vanish as ω goes to zero. Therefore, the terms $\pm\omega$ may be dropped from the energy denominators without altering the resulting correction to first order in ω. Thus

$$\epsilon_n = E_o + \omega(n+\tfrac{1}{2}) - \omega(n+\tfrac{1}{2})\, 4 \sum_{|E\rangle} \frac{|\langle E|W_3|E_o\rangle|^2}{E - E_o} + O(\omega^2) \,. \tag{A16}$$

which is of the form of (A6) where E_o is the polaron self-energy, and where μ is given by (A13). As was required to be shown, μ in (A6) is given by the same expression (A13) which was previously found for μ according to the definition (A7).

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